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STUDIES OF ANISOTROPIC PERMEABILITY WITH APPLICATIONS TO WATER REMOVAL IN FIBROUS WEBS
PART 2. RELATIONSHIP BETWEEN WATER REMOVAL AND PERMEABILITY, AND ADDITIONAL FACTORS AFFECTING PERMEABILITY.

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Studies of Anisotropic Permeability with Applications to Water Removal in Fibrous Webs

Part 2. Relationship Between Water Removal and Permeability, and Additional Factors Affecting Permeability.

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ABSTRACT

An extensive set of data for in-plane and transverse permeability of paper to water has been obtained. In Part I, we discussed experimental methods and presented data on anisotropic permeability in paper, as well as the relation between freeness and Darcian permeability. We found high anisotropy, with in-plane permeability ranging from 2 to 40 times as great as z-direction permeability.

In Part II, we present additional data on the effect of water removal on sheet permeability, including the effect of recycling. While drying a wet sheet of never-dried fibers to above 70% solids will cause a permanent increase in the permeability of the fibers from hornification, lower levels of water removal by wet pressing (or mild blotting and air drying) can also increase sheet permeability. This change is not due to true hornification, but may represent a temporary collapse of fibrils and rearrangement of fines by capillary forces.

We will also present data on the effect of fines content and the role that basis weight can play in measured sheet permeability. Finally, we discuss the nature of anisotropic permeability, the factors that cause it, and the implications it has for papermaking processes.

INTRODUCTION

In Part I, we outlined experimental methods for determining the in-plane and transverse permeability to water of saturated paper disks under various degrees of compression. Measurements of flow through the sheet are converted to yield curves of Darcian permeability as a function of sheet porosity (nonsolid volume fraction). We showed that in-plane permeability is substantially higher than the transverse or z-direction permeability at the same degree of compression (same porosity). The high flow anisotropy of paper can be important in wet press nips and other papermaking applications in which multidimensional flow occurs.

We now present further results from our study on permeability effects in paper.

FURTHER RESULTS

The Effect of Water Removal on Permeability

As a sheet is dried to a critical level over roughly 70% solids (1), the process of "hornification" occurs as capillary and other surface forces close many small pores, partially through bonding between microfibrils as water is removed. The closed pores are no longer open to water, permanently diminishing the swelling capacity of the fiber and reducing its swollen volume and its swollen surface area. As a result, when a dried sheet is resaturated or when dried fibers are reslushed and formed into a sheet again, less water is associated with the cellulose and there can be more space between the fibers open to water flow. As a result, permeability can increase.

While water removal is affected strongly by sheet permeability, we have found that the process of water removal at low solids levels (<50% solids) also changes the permeability of the sheet even after it is resaturated. Certainly compression of the sheet lowers permeability by reducing the pore volume, but at a given degree of compression or sheet porosity, a sheet made from never-dried fibers that has experienced previous water removal (typically over 30% solids) by pressing or blotting and air drying can exhibit higher permeability than a similar sheet at the same degree of compression that has been nearly saturated since formation.

In Figure 1 we show changes in the transverse permeability of saturated 135 gsm bleached softwood sheets which have been mechanically pressed to 25% or 50% solids or oven-dried to nearly 100% solids prior to resaturation. In Figure 2 we show similar results in unbleached softwood sheets pressed to different solids levels before resaturation. A change in pore structure is evident as water is progressively removed from the fibers. Partial water removal increases the permeability of the subsequently resaturated sheet. For example, the water removed in the first press may increase the permeability of the sheet when it is resaturated by compression in the second nip, although this may be offset by the increased density of the sheet.

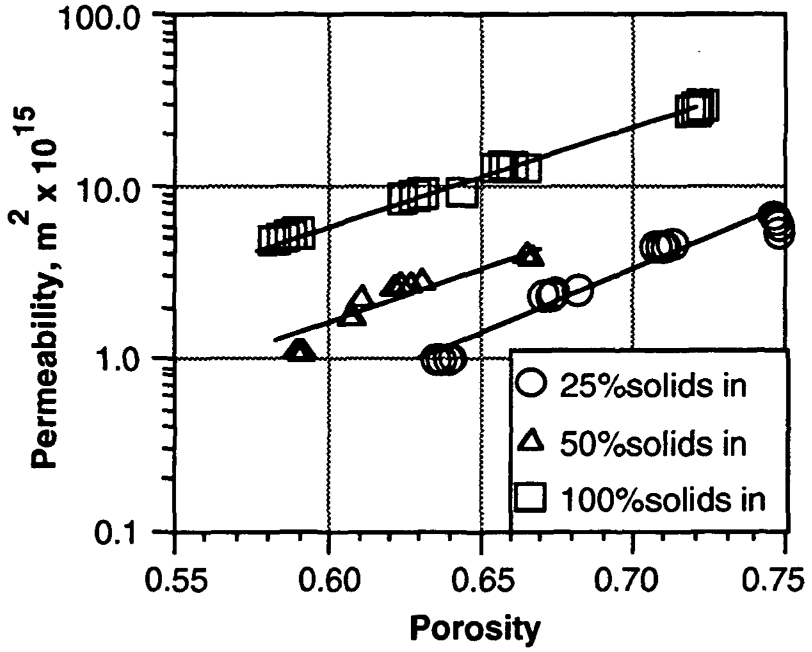

Figure 1. Effect of pressing and drying on the transverse permeability of a 135 gsm bleached softwood sheet.

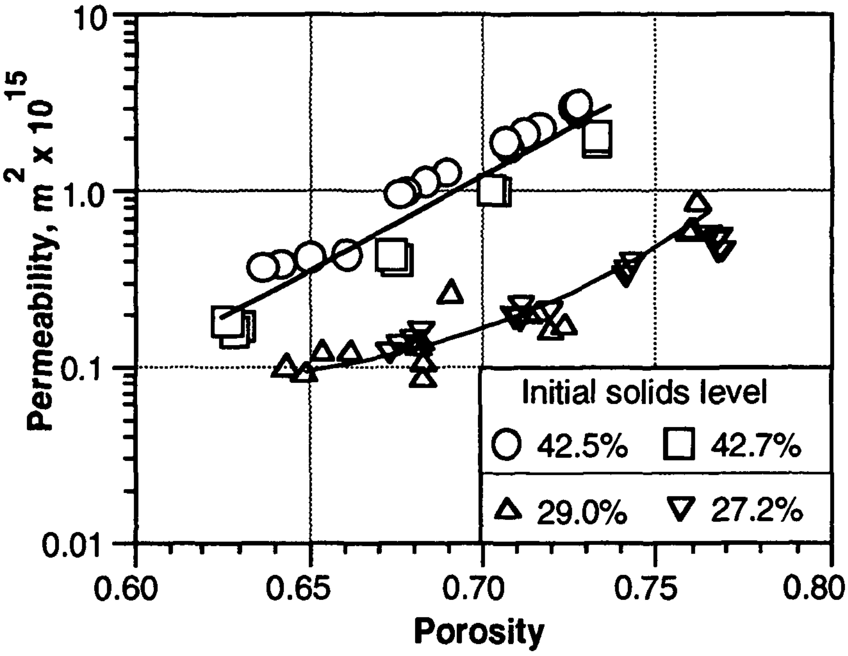

Figure 2. Effect of partial water removal through pressing on the transverse permeability of subsequently resaturated sheets of 200 gsm unbleached southern softwood kraft, 550 CSF.

Hornification and its effect on permeability is well known for recycled fibers (completely dried and reslushed), but the possibility of hornification during wet pressing operations is less well known. Washburn and Buchanan (2) noted that progressive water removal by pressing transforms the paper from a structure with loose fibrils to a compacted structure with collapsed fibrils and collapsed lamellae. Carlsson (3) reported a slight decrease in water retention values for fibers pressed to 65% solids versus 45% solids. Carlsson et al. (4) also noted that sheets which had been previously compressed to a high solids content had a higher permeability. The authors imply that it is the compression and not the partial drying of fibers that cause this effect. For the range of compression applied to sheets in this study, we have not observed changes in the permeability-porosity relationship due to compression as long as the sheet stayed saturated (i.e., water was added during expansion).

Recycled fibers. Figure 3 compares the effect of recycling (drying, reslushing, followed by handsheet forming on a British handsheet mold) with transverse permeability changes caused by pressing and drying of a sheet. A sheet made from the recycled fibers shows about the same permeability as a sheet made from never-dried fibers which has been pressed to 50% solids prior to resaturation. The recycled fibers are expected to have a lower permeability than a sheet which was completely dried because the process of recycling results in some fiber breakage, producing more fines. Furthermore, the enlarged interfiber pores caused by drying in a sheet are locked in place when the sheet is resaturated, whereas when the sheet is disintegrated to form a new sheet, the fibers are freed to possibly form a relatively tighter structure.

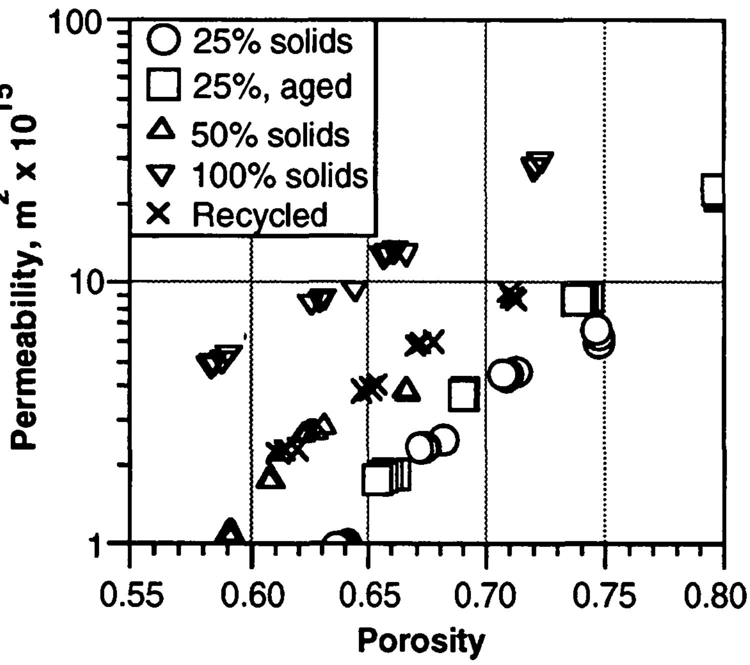

Figure 3. Comparison of permeability in a sheet of recycled fibers with partially dried sheets. Sheets are 135 gsm bleached softwood kraft, initially 715 CSF.

Partial Hornification? If the increase in permeability with partial water removal were due to the onset of hornification, a permanent change in fiber properties would be expected. To examine this possibility, further experiments were carried out. We formed a variety of sheets (all near 200 gsm) from a never-dried, unrefined, unbleached kraft pulp of southern softwood, following the program shown in Figure 4. One set of handsheets were kept fully saturated (below 25% solids), one set was repeatedly but gently couched and blotted to achieve a solids level of 40%, and other sets were air dried to 85% solids or oven dried at 108°C to essentially 100% solids. Permeability measurements were made for representative sheets from each set of handsheets; the sheets were resaturated prior to measurement. The various sets of handsheets (minus the 3-inch disks that were cut out for permeability measurements) were then reslushed and used to form handsheets again for further permeability measurements.

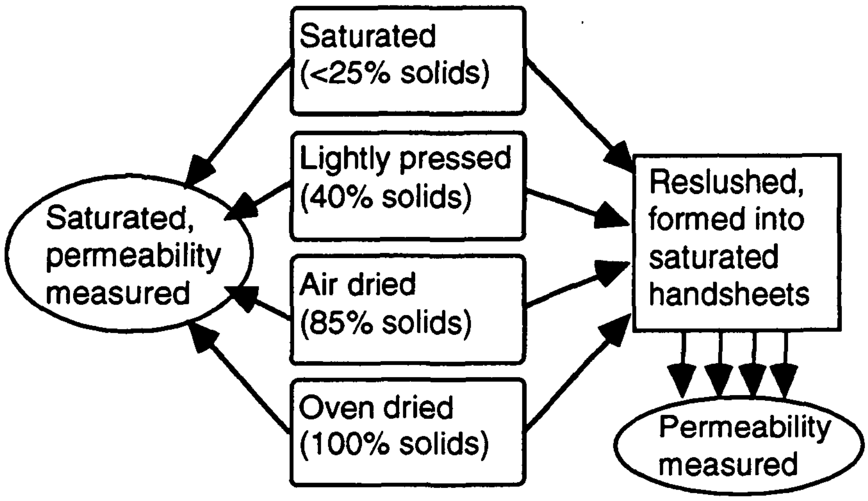

Figure 4. Outline of sheet preparation procedures for measurements of water removal effects on permeability.

Permeability results in the original handsheets, corresponding to the left side of Figure 4, are presented in Figure 5. The sheets that had reached 40% solids levels prior to resaturation had higher permeability than the sheets that had remained saturated. Sheets dried to higher solids levels had even higher permeability.

Figure 6 shows results for sheets that had been treated and reslushed, corresponding to the right side of Figure 4. When reslushed and formed into saturated handsheets again, the sheets that had been pressed to 40% solids were not significantly different in permeability than the reformed handsheets made from never-dried sheets. (The permeability of the reslushed, never dried sheets was higher than in the original handsheets from which they were formed, presumably due to a loss in fines in the handsheet forming process.) Yet the fibers which had been completely dried displayed a significantly higher permeability than the reformed handsheets from never-dried pulp, for hornification - a largely irreversible effect - had occurred. A permanent change in the fiber properties did not occur in the sheets pressed to 40%. Instead, we attribute the increase in permeability from "moderate" water removal to a change in the pore geometry due to capillary forces - e.g., from a reversible collapsing of fibrils and fines towards the fibers, clearing or creating pore spaces for water flow upon subsequent resaturation.

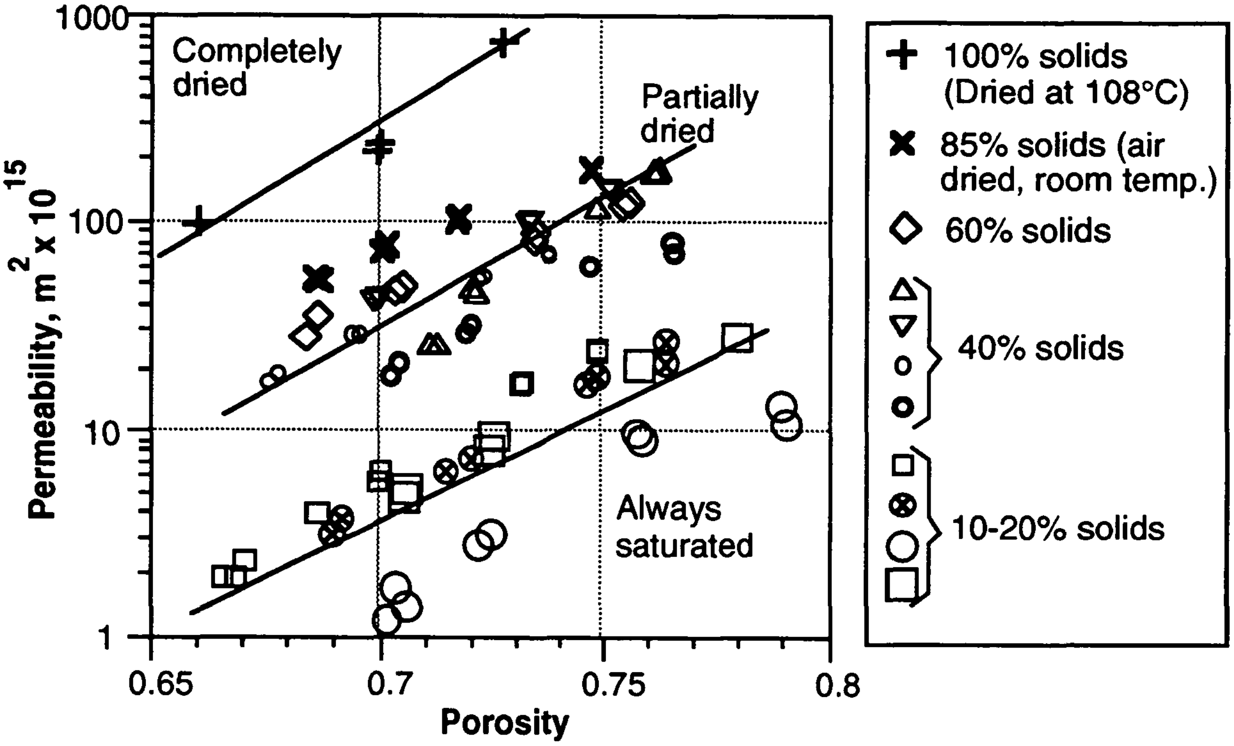

Figure 5. Permeability data for an unbleached softwood furnish showing the effect of water removal. Solids values represent maximum sheet dryness prior to resaturation and permeability measurement.

Based on the permeability changes observed in reslushed handsheets, irreversible swelling loss does not occur in this pulp until the sheet is dried above 85% solids. Our measurements reflect the change in macroscopic pore structure in a fiber mat and may not be sensitive to pore closures that do not significantly affect the interfiber pore space. We expect hornification to commence near 70% solids, but the pores that are irreversibly closed at this point may not have a strong enough effect on fiber stiffness or bulk swelling to change the permeability-porosity relationship. A more thorough discussion of the fiber-water interactions we have observed in water removal and recycling processes will be treated in a future paper.

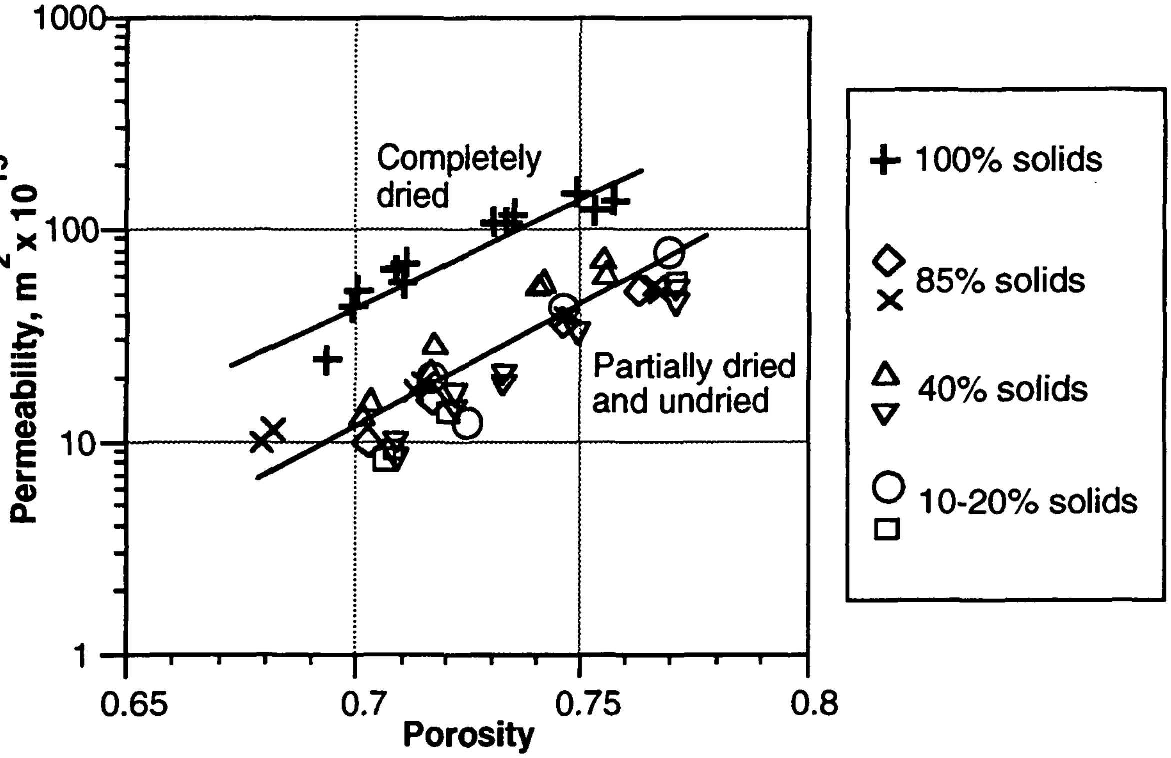

Figure 6. Permeability data for an unbleached softwood furnish showing the effect of water removal on sheets that are subsequently slushed and reformed into saturated handsheets. Solids values represent maximum sheet dryness prior to slushing.

Fines Content

Fines are known to have a significant effect on sheet permeability. This is demonstrated in Figure 7, where we compare permeability in sheets from classified and unclassified pulp. An increase in permeability by a factor of 10 was caused by removal of fines. A Bauer-McNett classifier was used.

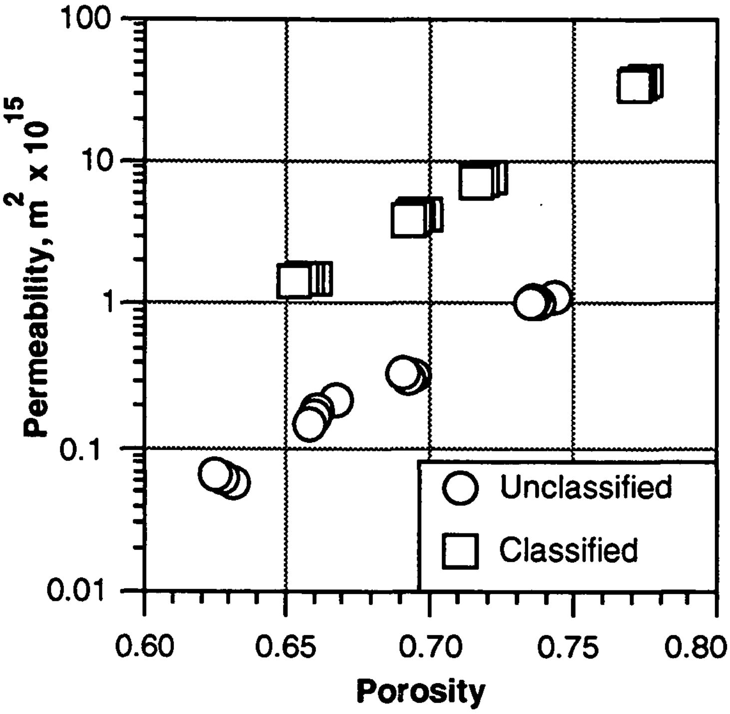

Figure 7. Effect of classification on permeability in 135 gsm sheets of bleached softwood, 654 CSF.

Basis Weight Variation

As noted previously, Ellis (5) reported that low-basis weight sheets tended to have higher permeabilities than heavier sheets formed from the same pulp. A similar effect was observed in this study for transverse permeability. Figure 8 shows data for several basis weights of bleached softwood pulp. The lightest sheet, 135 gsm, has a much higher permeability than the other sheets. This effect was consistently observed in this study. For example, Figure 9 shows the same trend in hardwood fibers.

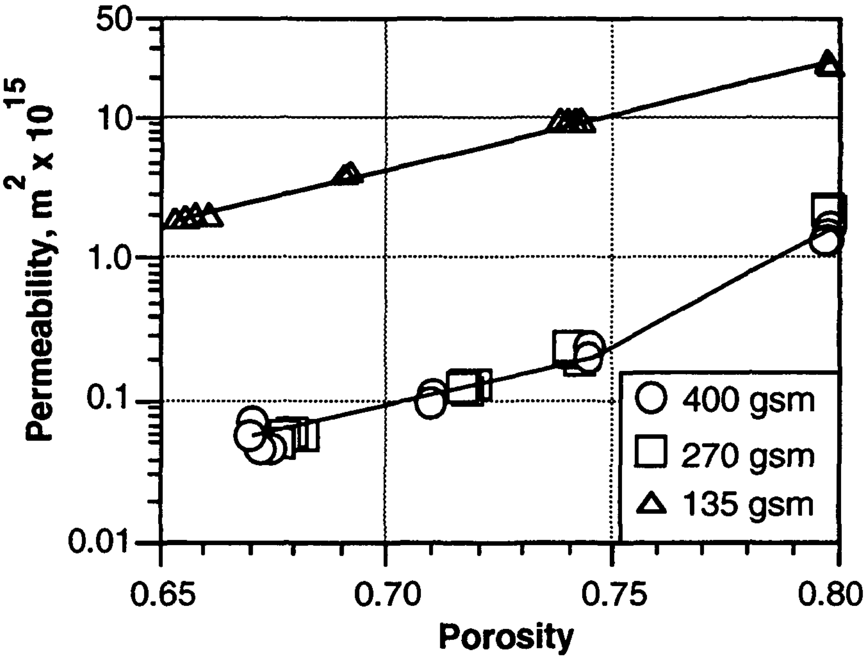

Figure 8. Basis weight effect in bleached softwood sheets.

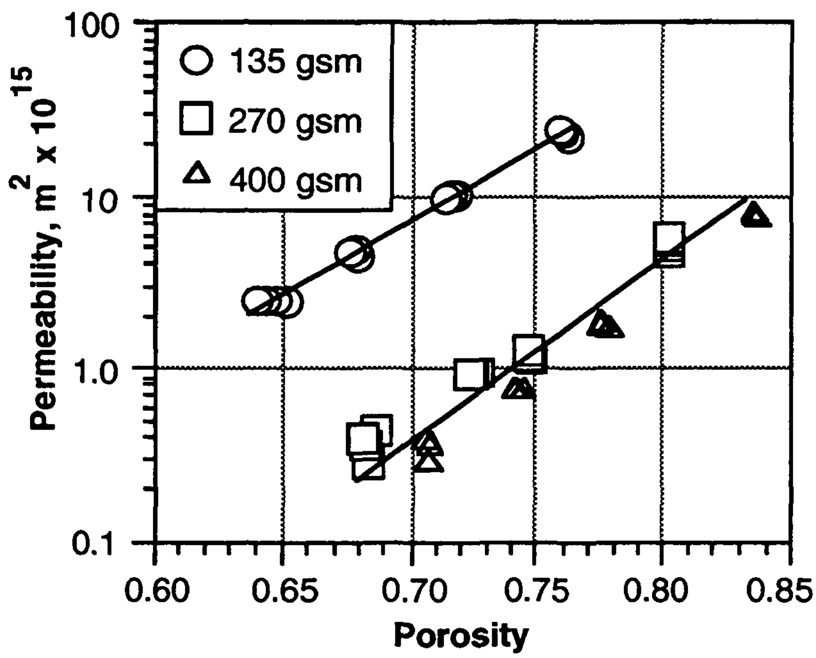

Figure 9. Basis weight effect in bleached hardwood sheets, CSF 567.

To explain the basis weight effect, it was hypothesized that fines distribution may play a role. According to this hypothesis, fines in heavier sheets may accumulate near the flow-exiting side of the sheet during handsheet formation, creating a low-permeability zone that reduced the apparent permeability of the sheet. In lightweight sheets, however, the smaller quantity of fines might not be sufficient to create a low-permeability layer. This hypothesis was rejected, however, after testing for basis-weight effects in classified pulp. Figure 10 shows the same basis-weight effect in classified bleached softwood pulp.

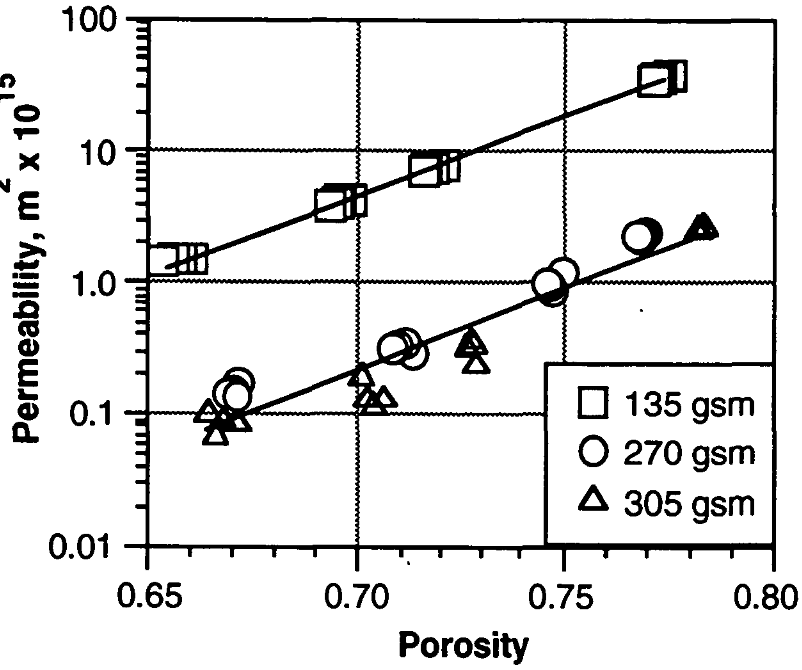

Figure 10. Effect of basis weight for sheets of classified bleached softwood pulp, 654 CSF.

Ellis had hypothesized that the basis-weight effect was due to experimental artifacts caused by the interaction of his sheets with the rough porous surface that supported the sheets during permeability measurement. In our experiments, the smoothness of the felts with respect to the sheet seemed easily sufficient to prevent such an artifact. It appeared that the high permeability was real.

A possible explanation for the effect came during examination of micrographs showing the structure of the sheets. We found occasional "macropores" in the sheets that could enhance flow in the z-direction over a short range. An example is shown in Figure 11, where an electron micrograph of a 270 gsm sheet has been modified with computerized color fills to show several regions of interconnected pores traversing the z-direction. The macropores may represent boundaries between flocs or aggregates that occurred during formation. Handsheet formation employs a low-consistency stock to prevent significant flocculation, so floc formation in the suspension may not be the proper explanation. The macropores could be due to sheet disruption by the flow during formation or by couching and pressing of the sheet (or may have been created during drying, in which case they would not help explain our observed basis-weight effect on permeability). This length scale of these pores approaches the thickness of the lightweight sheets but is less than the thickness of the heavier sheets. As a result, these pores could conduct a significant amount of water through a light sheet, where they may nearly pass from one end of the sheet to the other. In heavier sheets, however, these random, large pores do not transverse the majority of the sheet and thus are "averaged out" with the rest of the small pores in the sheet, creating an average permeability that no longer changes much with increasing thickness.

Figure 11. Electron micrograph of the cross-section of a 270 gsm bleached softwood sheet, modified with image analysis techniques to highlight several regions of interconnected pores.

Gren and Hedström (6) also note that nonuniform formation can cause an *increase in* measured permeability. Intrinsic nonuniformities in paper will thus cause an increased permeability until the sheet is thick enough for the nonuniformities to become small compared to the sheet thickness.

Further insight into the relation between basis weight and permeability comes from a study of air flow in paper by Knauf (7). Using a Gurley porosity tester, Darcian permeability to air in 60 gsm sheets was found to be significantly greater than in 200 gsm sheets. Effects due to surface roughness cannot explain this phenomenon.

A basis-weight effect was not seen in our lateral permeability measurements. In these tests, the flow path is about 3.7 cm through the plane of the paper, and the effects of random macropores are averaged out. However, if the measurement had been made using a much shorter flow path, still higher permeability values for in-plane flow may have been seen.

SOURCES OF ERROR

A variety of factors may affect the reliability of these results. We therefore examined reproducibility and possible experimental artifacts.

Testing for Artifacts

Inertial effects. Darcy's law is not valid when inertial effects become important in a porous medium. At high fluid velocities, the relationship between pressure drop and velocity becomes quadratic, not linear. We tested for inertial effects by measuring the apparent Darcian permeability in a sheet subject to three different pressure drops, with the maximum pressure drop well outside the range used in this study. All three data sets essentially fell on the same line, as shown in Figure 12, indicating that Darcy's law applied.

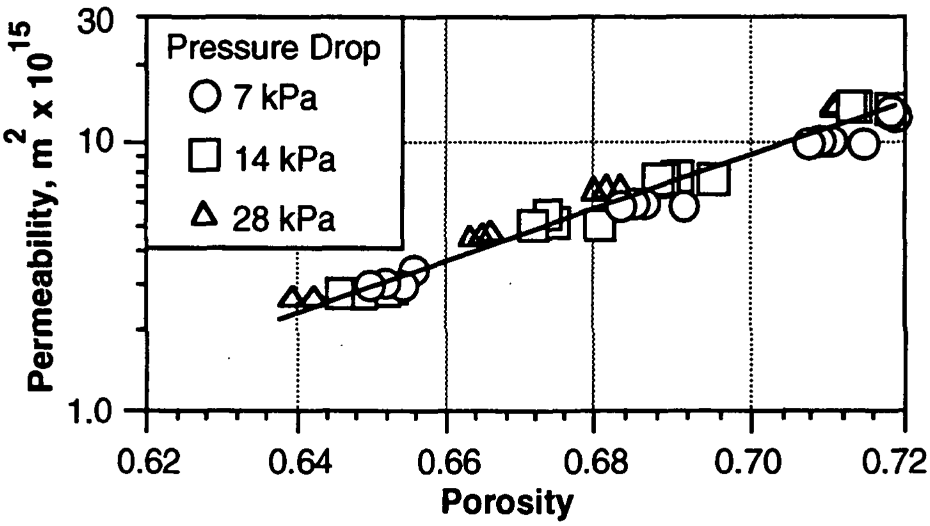

Figure 12. Measurements at different pressure drops to test for inertial effects. Sheet was bleached hardwood, 135 gsm, CSF 653.

Aeration. Another artifact which could be important relates to the effect of air bubbles on measured permeability. Some authors have suggested that careful deaeration of water and sheets is needed, otherwise air bubbles in the sheet could substantially reduce the measured permeability (3). In our testing, we kept the sheet saturated from the point of formation on to prevent intrusion of significant air, and generally used deaerated water. However, we wished to test what affect air bubble formation might have, for the use of pressurized air to pressurize our flow reservoir meant that the air content of the water would increase with time, possibly leading to artificial reduction in permeability during a run. However, when we measured transverse permeability using heavily aerated water, we could find no evidence of a permeability decrease due to air. In previous work we also found no evidence that deaeration of a saturated sheet had an affect on permeability. This is shown in Figure 13, where we compare lateral permeability measurements in four sheets of West Coast unbleached sulfite pulp, two of which had been deaerated for several hours under vacuum before making the measurement. At least for the procedures and samples of this study, permeability reduction due to air is not an issue.

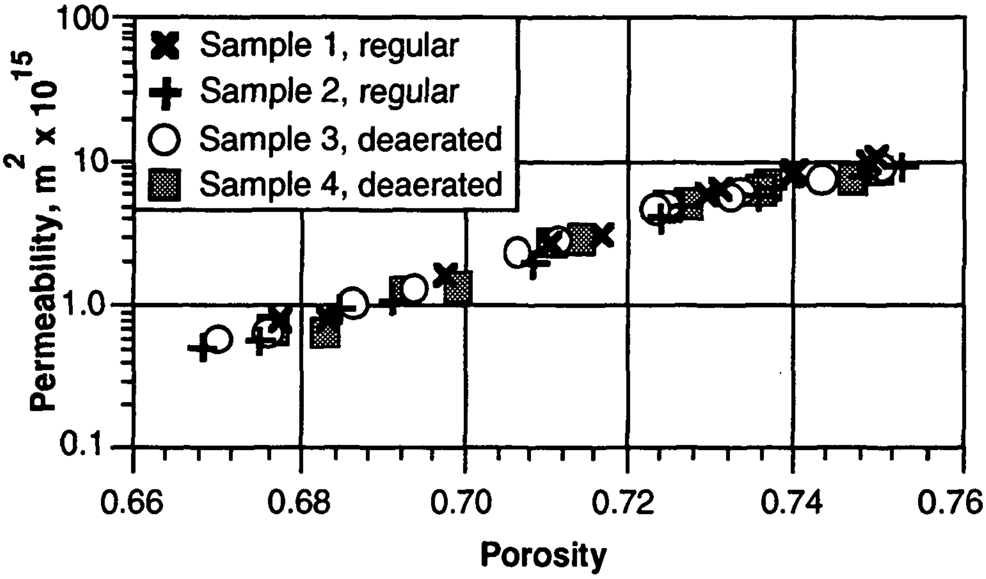

Figure 13. Effect of sheet deaeration on lateral permeability measurements of 200-gsm sheets of West Coast sulfite pulp.

Pulp storage time. We examined the effect of storage on the permeability of the pulp. The pulp was preserved with formaldehyde. After five weeks of cold storage, we could not detect a clear change in permeability for the softwood. The hardwood pulp showed some signs of microbial degradation, but a clear change in permeability was not detected.

Wire effect. The mesh size of the wire used in British handsheet mold was varied between 100 and 150 mesh. No effect of the wire could be detected between samples.

Reproducibility

Because of intrinsic nonuniformities and natural variation in sheet formation, as well as experimental error, measured permeability is subject to variance between samples and within a sample. For example, Figure 14 shows scatter between four data sets consisting of two separate 270 gsm sheets each tested twice. In addition to a difference between the two sheets, the replicate measurements on each sheet give slightly different results.

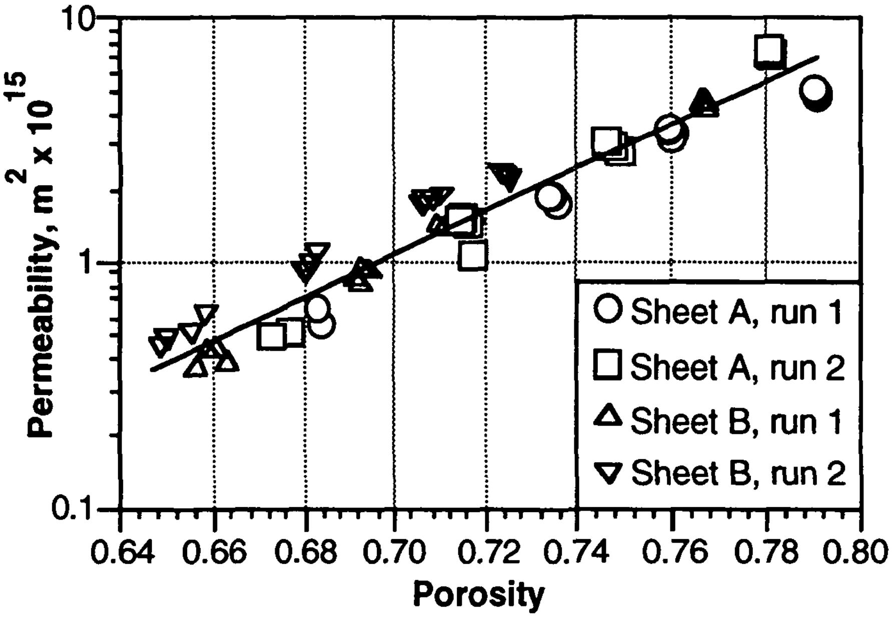

Figure 14. Reproducibility for 270 gsm sheets of unrefined, stored bleached hardwood.

Low basis weight sheets (135 gsm) were subject to additional scatter between samples, possibly due to the randomness of nonuniformities (macropores) which inflated sheet permeability, as discussed above. Figure 15 shows four runs for two samples of 135 gsm hardwood.

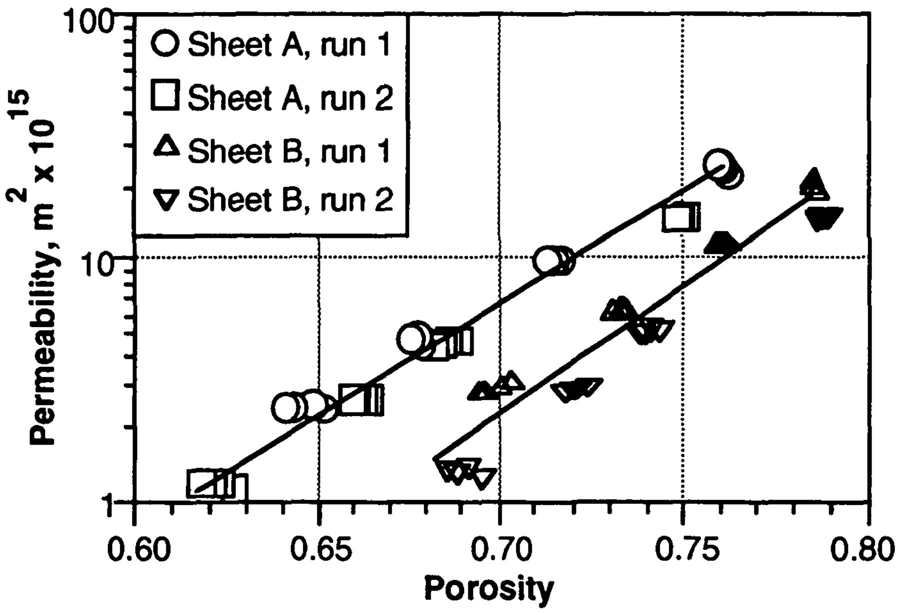

Figure 15. Reproducibility for 135 gsm sheets of unrefined, stored bleached hardwood.

One of the worst cases of scatter is shown in Figure 16, where we tested the heaviest sheets (400 gsm) made from PFI-refined, aged hardwood pulp. This pulp, having been subject to some microbial degradation and having been additionally refined in a PFI-mill, was extremely weak. The thick 400-gsm mat tended to ooze radially outward when compressed, and the sheet was easily disrupted. As a result, replicate measurements posed serious difficulties. Without refining, the fibers were somewhat stronger and seemed to yield less scatter, as shown in Figure 17.

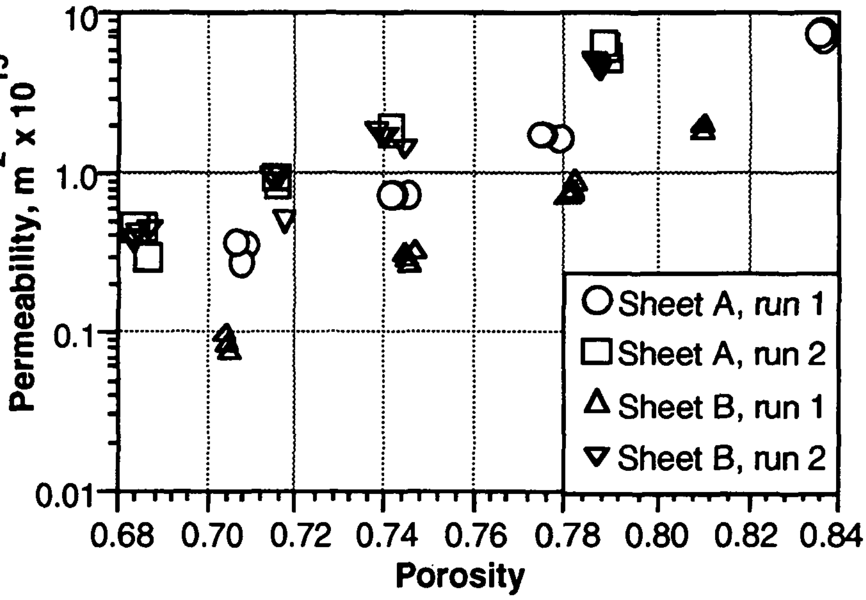

Figure 16. Large scatter in 400 gsm sheets of stored, PFI-refined bleached hardwood.

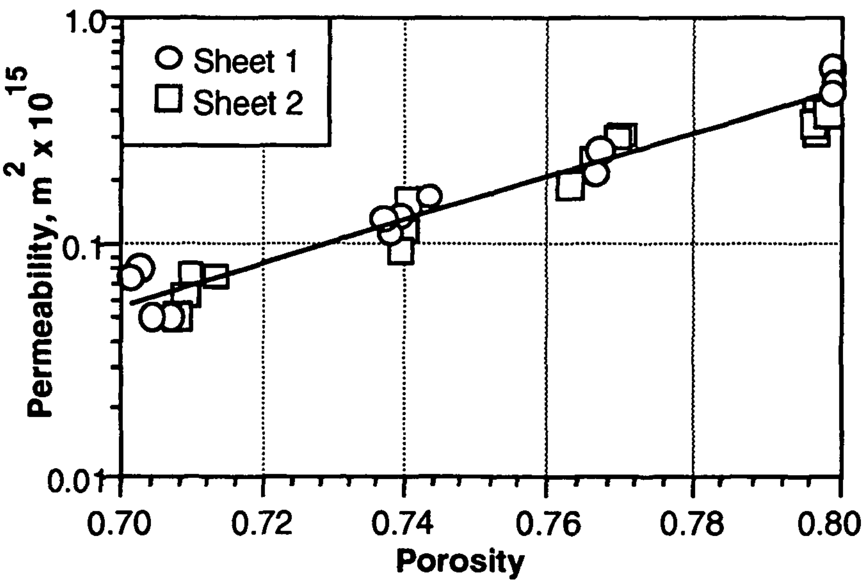

Figure 17. Reproducibility in 400 gsm sheets of stored, unrefined bleached hardwood.

DISCUSSION

Cause of Anisotropy

The anisotropy levels reported here are far beyond what has been predicted based on simple models of flow over aligned cylinders, where a ratio no greater than 2 is expected (e.g., see discussion in [8]). However, these models fail to describe the pore structure of consolidated paper. The flattened, fibrillated fibers in a compressed mat are much unlike an array of uniform cylinders. Theoretical work by Brown (9) for flow through arrays of elliptical fibers indicates that anisotropy values much greater than 2 may occur for fibers with flattened elliptical cross sections (high aspect ratios). This is consistent with the recent work of Hamlen and Scriven (10), who developed a computational model for three-dimensional pore structure and permeability in a web made of regular, deformable fibers. In one set of predictions, anisotropy ratios greater than 2 occur for high aspect ratio fibers. For example, an aspect ratio of 14 (height-to-width ratio of 0.07) was predicted to have an anisotropy ratio greater than 7 for a high linear fiber density.

In general, the combination of fiber geometry and planar fiber orientation creates an anisotropic flow network. Pores in the z-direction are continually interrupted by the broad projection of flattened fibers. Pores in the plane meander alongside fibers and over or under the thin projections of fibers. The final result is high in-plane permeability and low transverse permeability. The process of sheet formation also creates a pore structure with lower z-directional permeability than a random fiber network because of the "self-healing" effect in which random, open pores accommodate more flow and thus are quickly sealed as more fibers are deposited.

Changing pore structure during compression is likely to affect anisotropy. The change in anisotropy with compression is not yet well understood, although typical data sets in this study suggest a mild decrease in anisotropy with increasing compression (decreasing porosity). Hamlen and Scriven (10) predicted a strong decrease in anisotropy with compression, enough so that transverse permeability could exceed lateral permeability in sufficiently compressed webs. This possibility will be further explored in future work.

Dryness History, Recycling and Permeability

The elevated permeability observed in sheets at solids levels of 40% relative to sheets kept virtually saturated was not expected, although reasonable explanations can be suggested in terms of capillary forces causing increased openness of pores through a sheet. This result could be of significance in predicting the performance of press sections, for the permeability-porosity curve that characterizes a sheet may be altered if the solids level exceeds 40%. This might be important in impulse drying, for example, where high sheet permeability is desirable. Indeed, recent work in impulse drying has shown excellent water removal results when using sheets that have been pressed above 40% solids (11). Of course, many factors other than sheet permeability are involved in impulse drying performance.

Recycled fibers have the potential for high permeability and thus good pressing and drying performance. However, fines from the pulping process and other contaminants may reduce sheet permeability in industrial processes to levels below that of virgin fibers. If the secondary fibers are refined to regain the strength of the virgin fibers, the permeability may be substantially reduced. We are currently involved in exploratory research to find ways to preserve the high permeability of secondary fibers while obtaining good sheet strength.

Macropores and Basis Weight Variation

Based on measurements and micrographs, macropores between fiber aggregates may contribute substantially to the measured permeability of a sheet. In sheets too thin for these pores to be averaged out, a significant inflation in measured transverse permeability may occur. This effect was not seen in lateral permeability measurements because a long flow path through the plane of the sheet was used.

Lateral permeability may be even higher than we measured for flow processes where the in-plane flow occurs across a small distance. In this case, macropores may then inflate the apparent permeability, as they presumably did in our measurements of transverse permeability in thin sheets. Indeed, in processes such as blade coating, where the in-plane pressure gradient under the blade has a very small length scale, nonuniformities or macropores may play a major role in the penetration of coating into the paper. This is consistent with phenomena observed by Windle et al. (12).

An important implication of this finding is that moderate to low basis weight sheets may need to be treated as heterogeneous media rather than uniform porous media. Analysis of transport phenomena in processes like wet pressing, blade coating, and drying may be incorrect if the assumption of homogeneity is made, as it almost always is. There may be a need to consider complex stochastic transport models, which treat the porous medium as having a statistical distribution of pore sizes.

CONCLUSIONS

Based on the data presented above, the following conclusions are offered:

- In-plane permeability in paper can be higher than the transverse permeability by factors on the order of 2-40.

- For pressing and drying operations, permeability is a more useful measure of water removal behavior than freeness. Both in-plane and transverse permeability may be important in pressing operations.

- Partial water removal through pressing can change the pore structure in sheets, increasing the permeability. The effect does not appear to be true hornification.

- Low basis weight sheets may have higher than expected transverse permeability because of macropores between fiber aggregates, even in sheets formed from low-consistency slurries.

- The presence of heterogeneities such as macropores in paper may invalidate common assumptions used in analyzing transport processes in paper. Stochastic models may be needed in some cases.

Many issues deserve future study in order to better understand the water removal behavior of fibrous webs in real processes. In future work we will seek to firmly establish the change in anisotropy with compression, testing the computational prediction of Hamlen and Scriven (10) that the nature of the anisotropy can reverse at high compressive loads. The effect of chemical additives or filler particles on the anisotropy of fibrous materials is worthy of study. Permeability to two-phase flows in fibrous structures is another important and largely unexplored area. Gas-liquid flows in fibrous media occur in both papermaking and nonwovens applications. In future work we intend to measure the anisotropic relative permeability behavior of fibrous media in water-air and water-steam flows.

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